SOME PROPERTIES OF RADIOACTIVE FALLOUT: SURFACE DEVONATION COULOMB C

Final Report
September 1969

Contract No. DAHC20-69-C-0142 OCD Work Unit 3119A

URS RESEARCH COMPANY



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by

Carl F. Miller
URS RESEARCH COMPANY
155 Bovet Road
San Mateo, California 94402

for

OFFICE OF CIVIL DEFENSE
Office of the Secretary of the Army
Department of the Army
Washington, D.C. 20310

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SUMMARY

Data on the ionization-rate decay, activity-particle size distributions, and exposure rate-conversion coefficient for soil and filter-collected samples of fallout from Shot Coulomb C have been presented. Radiochemical analyses of selected samples permitted the correlation of the results in terms of equivalent fission levels relative to the Mo-99 content of the samples. The gross fractionation ratio relative to normal U-235 fission products was generally lower than observed for the fallout from larger yield explosions at all times from 240 to 6,000 hours after detonation. Individual radionuclide radiochemical "R" values were determined for Sr-89, Zr-95, Te-132, and Ba-140. No soil induced activities were detected to be present in significant amount at the time the analyses were carried out at around H + 270.

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INTRODUCTION

The surface detonation named Coulomb C was part of the PLUMBBOB weapons test series conducted at the Nevada Test Site in the summer of 1957; this surface detonation has been assigned a detonation yield of 0.5 kT. In this report, data on the decay of the fallout on activity-particle size distributions, on radiochemical analyses, and on the ionization rate conversion factor are reported.

No fallout collection and analysis program was sponsored for this event. The author and Mr. George T. Anton were visiting the test site for another purpose at the time and upon learning of the fallout pattern from this detonation, obtained permission from the site director to enter the area along with Mr. William J. Brady of the Reynolds Electric Company Radiological Safety Office for the purpose of taking some surface soil samples. In addition, a vacuum filter unit was used to pick up particles from the surface of a macadam road which crossed the fallout pattern. The samples were sent to the U.S. Naval Radiological Defense Laboratory, San Francisco, California, for radiochemical analyses, gross assay of radioactive content, sieve analysis, and measurement of the decay rates.

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RADIOCHEMICAL AND ASSAY RESULTS

Six soil samples were taken, each covering a square foot in area and about 1-inch in depth; three filter samples were taken at three points along the road crossing the pattern. The first two soil samples were taken on the hotline of the fallout pattern at a distance of 0.4 miles from ground zero where the exposure rate at 3 feet above the surface was measured to be 4 R/hr at 48 hours after detonation. The second set of two soil samples were taken on the hotline at a distance of 0.5 miles from ground zero where the exposure rate was 3.5 R/hr at 48 hours after detonation. And the third set of two soil samples were taken on the hotline at a distance of 1.8 miles from ground zero where the exposure rate was 660 mR/hr at 48 hours after detonation. The distance from ground zero to the road from which the vacuum filter samples were obtained was about two miles.

The gross radioactive content of all the samples was measured in the USNRDL Doghouse counter^{2,3} and then two of the soil samples were aliquoted for radiochemical analysis and decay measurements. Only one of the filter-collected samples contained sufficient activity for similar analyses. One of the soil samples was used to measure the decay rate in the Doghouse counter. The remainder were subjected to sieve analysis. The sample designations, relative gross activity corrected to 240 hours after detonation, and disposition of each sample are summarized in Table 1. The results of the radiochemical analyses of the aliquoted samples are given in Table 2. The i_d and i_g values for the aliquots indicate that some difficulties occurred in obtaining good separation of the Mo-99 from the soil samples (probably due to the bulk of foreign substances present in the solutions). The C9 sample analysis was done in triplicate and had much less inactive soil present; the i_g value from this sample was therefore used to derive i_g values for the decay samples and as a basis for estimating the a_g values for all the samples.

^{*} The analyses were carried out by D. Love, D. Sam, P. Strom, M. Nucholls, and D. MacDonald in December, 1957 and were reported in a USNRDL memorandum to the author. by Paul Zigman on January 21, 1958.

Table 1 DESIGNATION, GROSS ACTIVITY, AND DISPOSITION OF COULOMB C SAMPLES

Sample Designation	Type of Sample	a_{d} $\frac{\text{(cpm at } \text{H + 240)}}{\text{(cpm at }}$	Total Weight (gm)	Disposition
Cl	Soil	661,500	1,366.5	110.8 gm for analyses; remainder sieve analyzed
C2	Soil	933,200	-	not treated, retained for future analyses as needed
C3	Soi1	808,700	-	sieve analyzed
C4	Soil	801,900	1,097.8	62.1 gm for analyses; remainder sieve analyzed
C 5	Soil	161,700	-	sieve analyzed
C6	Soil	143,700	-	Doghouse decay
C7	Filter	1,077	-	not treated
C8	Filter	3,283	-	not treated
С9	Filter	55,030	-	entire sample subjected to radiochemical and other analyses

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Table 2
SUMMARY OF RADIOCHEMICAL ANALYSIS AND ASSAY RESULTS
ON COULOMB C FALLOUT SAMPLE ALIQUOTS

			Aliquot	Designat:	ion	
Quantity Measured	Cla	Clb	Clea	C4a	C4b ^a	C9 ^a
Weight (gm)	37.84	32.05	40.90	30.79	31.30	***
a (cpm at H + 240)	12,590	13,730	16,500	34,080	15,380	55,030
ag(10 ⁻⁹ ma at H + 240)	123	134	165	345	150	511 ^b
ag/ad(10 ⁻¹² ma/cpm)	9.77	9.76	10.00	10.12	9.75	9.29
a ₁ (10 ¹³ fissions) ^{c,d}	6,28	5.15	-	12.70	-	15.58 ^e
$i_d(10^{-9} cpm/f at H + 240)$	0.200	0.267	~	0.268	-	0.353
$i_g(10^{-20} ma/f at H + 240)$	0.196	0.260	-	0.272	~	0.328 ^e

- a Aliquots Clc, C4a, and 1/10 of the dissolved portion of sample C9 were used for decay measurements in the ion chamber.
- b a_g for the dissolved portion of the sample at H + 268 was 323×10^{-9} ma indicating that only about 0.708 of the activity was dissolved and retained in solution (a certain loss of volatile activities including the iodine isotopes 131 and 132 would be expected at this time after detonation); $a_g(268)$ for the original sample would have been 456×10^{-9} ma assuming a decay rate as for C4b.
- c Based on Mo-99 analysis.
- d Radiochemical "R" values reported were: 0.040 for Sr-89; 0.059 for Te-132; 0.23 for Ba-140 in Sample C9; and 0.51 for Zr-95 in sample C4a.
- e Analysis of solution in triplicate gave 1.2×10^{14} fissions for the portion dissolved; this would give an i_g value of 0.426×10^{-20} ma/fission at H + 240 which is too high. Assuming partial solution of the sample, a_f would be given by $(456/323) \times 1.2 \times 10^4$ or 1.69×10^{14} fissions. Using 0.328×10^{-20} ma/fission at H + 240 as derived for aliquot C4b in Table 3 gives an a_f value of 1.56×10^{14} fissions; the latter value was selected since it is apparent that at least some activities were lost in the dissolution process.

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The a_f value for the decay sample C9a, at 0.1 volume of the solution from C9 is 1.20×10^{13} fissions. Comparing the decay curve measurements in the time period of 2,000 to 6,000 hours after detonation for samples C1c and C9a gives a ratio of a_f (C1c) to a_f (C9a) of 4.14 and, in the time period of 400 to 1,300 hours after detonation for samples C4b and C1c, a ratio of a_f (C4b) to a_f (C1c) of 0.920. The estimated a_f values from these ratios are therefore 4.97×10¹³ fissions for sample C1c and 4.57×10¹³ fissions for sample C4b; the a_f and a_f values for the three samples at different times after detonation are given in Table 3. The data for aliquot C1c is plotted with time after detonation in Figure 1; the curve has the characteristic curvature to a steeper slope starting at 2,500 hours after detonation. The slope of the straight line portion of the curve from about 300 to 2,000 hours after detonation is -0.86.

The variation of the gross fractionation ratio, $\boldsymbol{r}_{\mbox{\scriptsize fp}},$ with time after detonation for each of the three aliquots is shown in Figure 2. The shape of the curves are similar to those given in Reference 2 for the fallout from the tower detonations Diablo and Shasta. The r_{fp} values for the dissolved portion of sample C9 are lower than those for the two soil samples until about 2,000 hours after detonation due to apparent losses during the dissolution process of such nuclides as I-131, I-132, Ru-103, Rh-105, and perhaps Te-132 all of which would be present in an unfractionated FP mixture in significant quantity at H + 240 to H + 270. At this time, about 27 percent of the gamma ray energy would be contributed by the Iodine isotopes and the Lanthanum isotopes would contribute about 51 percent at H + 240. Up to about 1,000 hours after detonation the effect on r_{fp} of losses in both the I and Ru(Rh) isotopes would be possible; after about 1,500 hours after detonation, effects on the value of r_{fp} could only be due to Ru(Rh) isotopes and these could persist at times longer than 6,000 hours after detonation (i.e., at times beyond the time at which the Zr(Nb) isotopes are the predominating contributors to i_r). The differences between the r_{fp} values for aliquot Clc and C4b are very small in the period of 240 to 1,270 hours after detonation.

With the decay data of Table 3, revised values of a $_{\hat{\mathbf{f}}}$ and i $_{\hat{\mathbf{d}}}$ for the various aliquots are as follows:

Table 3 $\label{table 3} \mbox{SUMMARY OF IONIZATION-RATE DECAY MEASUREMENTS} \\ \mbox{AND $r_{\tt fp}$ VALUES OF COULOMB C FALLOUT SAMPLES}^a$

ig(10⁻²⁰ma/fission)

						r.	
Time After	_			Reference for		fp	
Detonation		ole Alic		Unfractionated		le Aliq	
(hours)	Clc	C4b	C9a	U-235 FP	Clc	C4b	C9a
240	0.332	0,328		1.00	0,332	0.328	
242	0.330			0.995	0.332		
243	•	0.324		0,985	•	0.329	
245	0.326			0.960	0.340		
260	0.310	0.304		0.915	0.339	0.332	
268	0.300	0.293	0.269	0.885	0.339		0.304
290	0.278	0.271		0.808	0.344	0.335	
319	0.252	0.247		0.724	0.348	0.341	
331	0.242	0.241		0.700	0.346	0.344	
334			0,239	0.690			0.346
356	0.229	0.225	0.227	0.642	0.357	0.350	0.354
411	0.205	0.204	0.198	0.550	0.373	0.371	0.360
506	0,172	0.170	0.163	0.440	0.391	0.386	0,370
525	0.166	0.165	0.158	0,422	0.393	0.391	0.374
598	0.152	0.153	0,144	0.376	0.404	0.407	0.383
602	0,148	0.148	0,142	0.358	0.413	0,413	0.397
672	0.134	0.134	0.128	0,314	0.427	0,427	0.408
722	0.125	0.126	0.119	0.285	0.439	0.442	0.418
770	0.119	0.120	0.110	0.260	0.458	0.462	0.423
843	0.110	0.111	0,101	0.229	0.480	0.485	0.441
908	0.103	0.103	(1)950	0.206	0.500	0.500	0.461
937	(1)998	0.100	(1)933	0.197	0,507	0.508	0.474
1,008	(1)938	(1)943	(1)883	0.180	0,521	0,524	0.491
1,080	(1)884	(1)880	(1)842	0.162	0.546	0.543	0.520
1,270	(1)757	(1)757	(1)749	0.127	0.596	0.596	0.590
1,610	(1)622		(1)608	(1)888	0.700		0.685
2,020			(1)487	(1)659			0.739
2,050	(1)515			(1)640	0,805		
2,420			(1)429	(1)528			0.812
2,460	(1)433			(1)518	0.836		
3,190	(1)316		(1)319	(1)375	0.843		0,851
3,910	(1)246			(1)286	0.860		
4,620	(1)185		(1)188	(1)211	0,877		0.891
4,800			(1)173	(1)195			0.887
4,920	(1)164		(1)162	(1)185	0.886		0.876
5,640	(1)121		(1)121	(1)139	0.870		0.870
6,410	(2)884		(2)858	(1)104	0.850		0.825
7,100	(2)654		(2)628	(2)780	0.838		0.805
7,920	(2)468		(2)425	(2)570	0.821		0.746
8,660	(2)335			(2)430	0.779		
9,450	(2)242			(2)325	0.745		
10,120	(2)185			(2)262	9.706		
11,080	(2)129			(1)192	0.672		
12,280	(3)872			(1)135	0.646		

a Numbers in parentheses are the number of zeros between the decimal point and the first digit.

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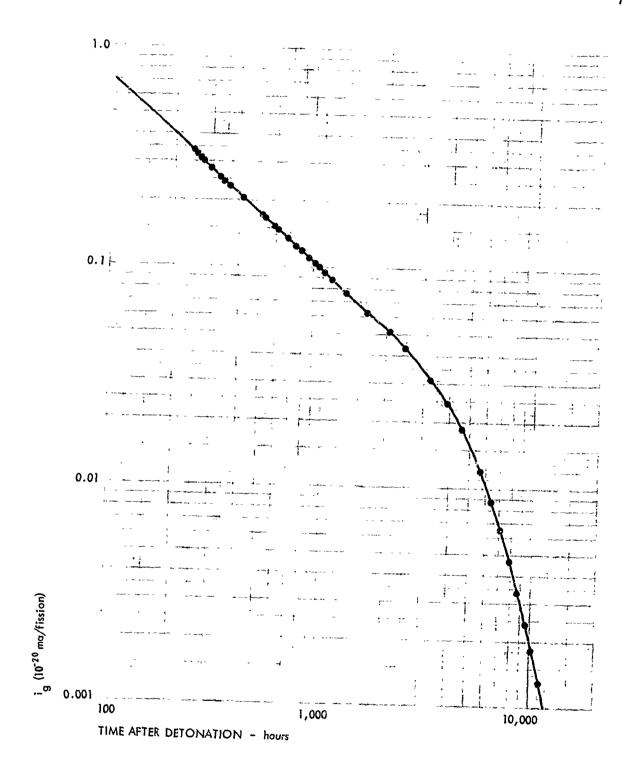


Figure 1. Ionization-Rate Decay of Aliquot Clc of Coulomb C Fallout Sample Cl.

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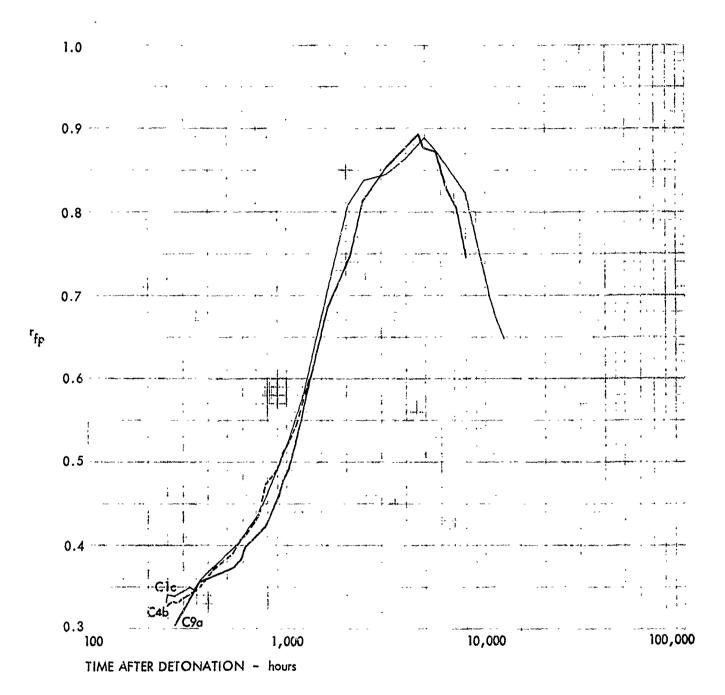


Figure 2. Variation of $r_{\mbox{fp}}$ with Time after Detonation for Aliquots of Coulomb C Fallout Samples.

Aliquot	a _f (1: ¹³ f)	i _d (10 ⁻⁹ cpm at H + 240/f)
Cla	3.705	0.340
Clb	4,036	0,340
Cle	4.970	0,332
C4a	10.52	0.324
C4b	4.573	0,336
C 9	15.58	0.353
Average	43	0.337

The Doghouse count-rate decay of Sample C6 is given along with the ratio of i_g to i_d in Table 4; the increase of this ratio with time from about 240 to 800 hours after detonation shows that the count-rate decay is more rapid in this time period than is the ionization-rate decay.

The average H + 240 value of i_d given above was used to calculate the equivalent fission contents and surface density of activity for the three soil sampling locations using the Doghouse count-rate data given in Table 1. The results and calculated values of K_S , the H + 1 exposure rate activity conversion coefficient, are given in Table 5; the average value of K_S for the last five samples (some of sample Cl was spilled in transfer) is 864 R/hr at 1 hr per KT/sq mi. The K_S values, although in the proper range, may be in error by as much as 15 percent due to the errors involved in estimating the decay correction factor (not considering other sources of error).

Table 4

DOCHOUSE COUNT-RATE DECAY OF SAMPLE C6

Time After	ie	ig/id
Detonation (hours)	(10^{-11}cpm/f)	(10 ⁻¹² ma/cpm)
240	33.7	9.79
244	32.9	9.88
300	24.9	10.8
320	21.7	11.5
332	20.4	11.8
357	18.5	12.3
410	15.4	13.2
434	14.2	13.7
504	12.0	14.2
600	10.1	15.0
672	8.92	15.0
770	7,75	15.5
844	7.14	15.5
900	6.81	15.1
938	6.57	15,2
1,007	6.10	15.4

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Table 5

CALCULATED VALUES OF OBSERVED EXPOSURE RATE-ACTIVITY CONVERSION

COEFFICIENTS FOR COULOMB C FALLOUT SAMPLE LOCATIONS

			Sample	Number	_	
Quantity	<u>C1</u>	C2,		C4	C5	<u>C6</u>
r(mi) ^a	0.4	0.4	0.5	0.5	1.8	1.8
Λ _f (10 ¹⁵ fissions/sq ft)	1.96	2.77	2.40	2.38	0.479	0.426
I(R/hr at 48 hr)	4.0	4.0	3.5	3.5	0.66	0.66
I _s (R/hr at 1 hr) ^b	450	450	400	400	75	75
$K'_{s}(10^{-13}R/hr at 1 hr)/(f/sq fc)$	2,30	1.62	1.67	1.68	1.57	1.76
K _S (R/hr at 1 hr)/(KT/sq mi)	1,200	840	870	870	820	920

a r is the distance from ground zero to sampling locations on the fallout pattern hot line.

b the decay correction factor was calculated using the assumption that the ratio of the $r_{\rm fp}$ curve for Coulomb C fallout to that for the Diablo/ shasta fallout remained the same at 1 and 48 hours after detonation as it is for 240 hours after detonation; this assumption gives $i_{\rm g}(1)=248\times10^{-28}$ ma/fission and $i_{\rm g}(48)=2.19\times10^{-20}$ ma/fission which results in a decay correction from 48 to 1 hour of 113. A $t^{-1.2}$ decay correction factor would be 104 from 48 to 1 hour after detonation; the $r_{\rm fp}$ value associated with the indicated values of $I_{\rm S}$ is 0.230.

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ACTIVITY DISTRIBUTIONS FOR COULCMB C FALLOUT SAMPLES

Data on the sieve analysis of Samples C1, C4, and C5 were reported in memorandum form to the author by E. A. Schuert. The samples were drysieved and the sieve fractions were then assayed for gross activity with the Doghouse counting system at H + 357. Many spherical particles with diameters in the range of 506 microns were observed in Sample C1. These particles appeared to be melted silicates or silica; their general color was smoke grey but a small percentage were clear glassy particles.

The count-rate data in percentage form are given in Table 6 along with the results of a distribution curve analysis as shown in Figure 3. The samples appear to consist of two distributions. The small and large particle distributions from the samples at downwind distances of 0.4 and 0.5 miles were almost the same with a fallout particle diameter range of 600 to about 4,000 microns and a median diameter of 1,600 to 1,800 microns. At the downwind distance of 1.8 miles, the diameter range for the fallout distribution is estimated to be 190 to 850 microns with a median diameter of 415. The characteristic S-shaped distribution curves are obtained when the particle diameter limits are imposed. In the case of sample C5, the background or "small particle" distribution actually contains both smaller and larger particles than the fallout or "large particle" distribution.

^{*} Schuert, E. A., U.S. Naval Radiological Defense Laboratory memorandum to C. F. Miller, dated 30 December 1957.

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Table 6
SUMMARY OF SIEVE ANALYSIS OF COULOMB C FALLOUT SAMPLES

Diameter		Percentage of Gam	
(microns)	_ <u>C1</u> _	<u>C4</u>	C5
	1. Total Sample D	istribution	
> 2,000	100,00	100.00	100.00
840 - 2,000	66.96	59.08	99.97
590 - 840	15.06	13.59	95.43
420 - 590	5.54	4.16	82.31
300 - 420	3.84	2.80	51.11
250 - 300	1.45	1.48	12,19
150 - 250	1.45	1,47	7.64
105 - 150	0.35	0.34	0.94
75 - 105	0,16	0,16	0.49
< 75	0.072	0.052	0.13
	2. Background Dis	tribution ^a	
> 2,000	100.0	100.0	100.0
840 - 2,000	98.3	97.1	99.4
590 - 840	65.0	60.7	74.6
420 - 590	46.2	40.4	55.2
300 ~ 420	32.0	27.2	37.6
250 - 300	12.1	14.4	23.2
150 ~ 250	12,1	14,3	16.6
105 - 150	2.9	3.3	5,2
75 - 105	1.3	1.6	2.7
< 75	0.60	0.50	0.72
	3. Fallout Particle	Distribution	
> 2,000	100.0	100.0	100.0
840 - 2,000	62.7	54.7	100.0
590 - 840	8.25	8.18	100.0
420 - 590			88.2
300 - 420			54.1
250 - 300			9.76
150 - 250			5.67
d (migrans)	600	600	190
d _{min} (microns)	000	ΨOO	130
d ₅₀ (microns)	1,600	1,800	415
d (microns)	~ 4,000	~ 4,000	850

a About 12.0 percent of the total activity was with the background distribution for Sample C1; about 10.3 percent for Sample C4; and about 18.1 percent for Sample C5.

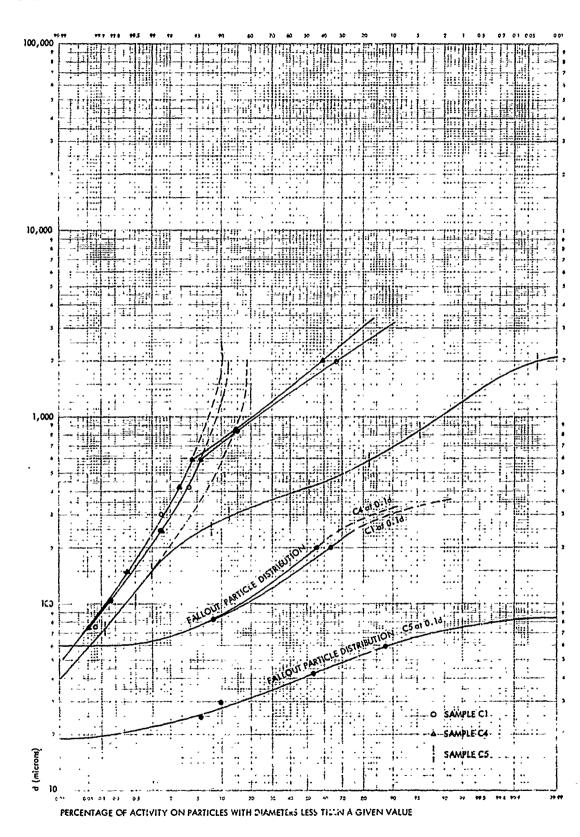


Figure 3. Activity Distribution of Soil Samples Taken from Three Different Locations on the Coulomb C Fallout Pattern.

SUMMARY AND CONCLUSIONS

Data on the ionization-rate decay, activity-particle size distributions, and exposure rate-conversion coefficient for soil and filter-collected samples of fallout from Shot Coulomb C have been presented. Radiochemical analyses of selected samples permitted the correlation of the results in terms of equivalent fission levels relative to the Mo-99 content of the samples. The gross fractionation ratio relative to normal U-235 fission products was generally lower than observed for the fallout from larger yield explosions at all times from 240 to 6,000 hours after detonation. Individual radionuclide radiochemical "R" values were determined for Sr-89, Zr-95, Te-132, and Ba-140. No soil induced activities were detected to be present in significant amount at the time the analyses were carried out at around H + 270.

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13. ABSTRACT		

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Unclassified
Security Classification